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RADIOACTIVE FALL-OUT IN RAIN IN THE CINCINNATI AREA

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Robert A. Taft Sanitary Engineering Center, Cincinnati, Ohio

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Radioactivity Fall-Out in Rain in the Cincinnati Area

by

J.S. Nader, A.S. Goldin and L.R. Setter

A study of the radioactivity of precipitation in the Cincinnati area was made for twelve months beginning March 1953. Seven samples of precipitation between March 3 and March 18 were tested to obtain the background level in rainfall. On March 19, two days after the first weapon test of the Spring Series, a sharp rise in the beta activity of rain occurred. The radioactivity of subsequent rains during the test period was usually of greater intensity and varied widely. Following the weapon tests the activity of the rain decreased to levels near that of background. All activity measured was far below the intensity considered to be hazardous. The more pertinent findings of this study are presented.

The rainfall, falling on a tray 0.54 square meters in area, was collected at 1014 Broadway, Cincinnati, Ohio, until October, when the sampler was moved to the roof of the Robert A. Taft Sanitary Engineering Center, at 4676 Columbia Parkway. At intervals between infrequent precipitation the collecting tray was flushed with water to remove soot-fall.

PROCEDURE

Rain accumulations were usually taken from the sampler between 8 and 9 A.M. on weekdays, and week-end rainfall was allowed to accumulate until Monday mornings. Several samples of snow were collected from relatively clean surfaces. The volume of rainfall was measured and the Weather Bureau reports were consulted for an estimation of the average precipitation in the Cincinnati area. When several samples were collected per day, the rainfall was prorated on the bases of sample collections.

An aliquot of rain was filtered through a membrane filter for an assay of radioactivity associated with suspended solids and the filtrate was evaporated, dried and counted for an assay of the "soluble" radioactivity (2). The suspended solids and dissolved solids were determined to estimate the self-absorption correction. This correction factor was determined from measurements of samples of various thickness prepared from an active rain.

The net counting rate was corrected for geometry, backscatter and self-absorption and reported in micro-microcuries per milliliter ($\mu\mu$c/ml). Generally the suspended solids radioactivity was counted on the day of sample collection and the filtrate or soluble activity was counted within 24 hours. The sum of these two values, uncorrected for decay, is reported as total beta radioactivity.

Most of the samples which had an appreciable activity were counted periodically for several weeks or until decay had reduced the count to approximately twice background.

RESULTS

The more pertinent results of these tests are presented in Table 1. The rains were arbitrarily grouped into nineteen sample series -- one covering the period before the weapons tests, eleven covering the periods following each of the eleven bomb bursts, and seven covering periods of one-half to two months each, subsequent to the bomb tests. The snow and rain samples prior to March 18, 1953 had a "background" radioactivity level of 0.03 to 0.08 μμc/ml. of total activity of which usually 50 to 100 percent was soluble.

During the weapon test period the radioactivity of the rain increased many fold. A maximum activity of 319 μμc/ml was observed in a small rain of 0.02 inches on April 29. This rain had a decay rate comparable to four-day-old fission product. When extrapolated to age three days the activity may be estimated to be 500 μμc/ml (0.0005 μc/ml).

Thirty percent of the total activity of this sample was soluble. According to the Atomic Energy Commission, "Safe concentrations of a mixture of fission products in drinking water depend upon the composition of the mixture and upon the period of time over which the water is used. It is estimated that water containing total fission product activity amounting to 0.005 microcurie per milliliter three days after the fission products were formed could be used safely for any period of time." The maximum fall-out of radioactivity from a single rain occurred on May 22 when 1.55 inches of rain containing 85.7 μμc/ml fell, resulting in a fall-out of 8.75 curies per square mile.

By August 4, or two months after the last test, some rains indicated a background level comparable to that before the weapon tests. However, six of the twelve rains tested in August, September and November and eight of the twenty rains tested in December, January and February had a radioactivity of two to three times the background level.

A study of the decay curves of rains as compared to the theoretical decay of fission product [3] indicates that the activity of a rain may be due to a bomb burst twice or thrice removed from the sample series. Typical examples of this are shown in Table 2, where the half-life of a specific rain sample of maximum activity in each of eleven sample series (Column 4) is compared to calculated half-lives for bomb fission products (Columns 5 to 15). The half-lives of rain and bomb material were measured from the date shown in column 3. The activity in the rains of the 1st, 2nd, 3rd, 5th, 7th and 9th sample series appears to be the result of the bomb burst immediately preceding the rains (see blocked values), whereas the remaining rains appear to be the result of earlier bursts. For example, the rain in the 4th sample series on April 13 appears to be due to the 2nd bomb burst on March 24. Activity from products formed in the sixth bomb burst on April 18 apparently was not observed in rains at Cincinnati.

For each sample series, the weighted average radioactivity of the rainfall, the weighted average percentage of soluble activity, and the total radioactivity which fell per square mile were calculated, and are presented in Table 3 and Figure 1.

Figure 1 shows graphically that from an average background level of 50 μμc per liter (log 1.7) of rain in sample series 0, the activity rose to a maximum of 80,600 μμc/l (log 4.9) for a sample series. Following the last bomb burst on June 4, the average activity in the rain decreased to within the range 30 to 80 μμc/l.

The percentage of soluble activity in the rain, which was 60 percent of the total activity before the spring bomb tests, fell to less than 30 percent during the tests. The soluble activity then increased to about 80 percent after the weapons test series.

Column 3 of Table 3 gives a rough estimate of the average age, in days, of the fission product in each sample series. From this age, and the theoretical decay curve (3), an estimate was made (Columns 8 and 9) of the radioactivity per square mile which would be left on June 10 and December 10, 1953. A summation of the fall-out per square mile for the period March 19 to June 10 (Series 1-11, inclusive) shows that, of the 13.5 curies of activity which fell per square mile, the residual on June 10 would be 2.4 curies. On December 10 the residual activity from the rains which fell from March 19 to July 25 would be 0.15 curies per square mile. In metric units, 2.4 curies/sq. mile is equivalent to 9×10^{-5} μc/cm^2, and 0.15 curies/sq. mile is equivalent to 6×10^{-6} μc/cm^2. Factors of runoff, percolation, etc. would obviously influence the distribution of the residual activity in the area.

A few samples of surface and cistern water were tested for radioactivity. A cistern water sample was tested on April 7 immediately after an eight day wet season of 0.86 inches rainfall containing an average activity of 2500 μμc/l. The roof water seeped through a wall of cinder blocks into the cistern. The cistern water activity was found to be only 29 μμc/l. On April 15, following an additional 0.46 inches of rain of comparable activity, another similar cistern water was tested. Its activity was 20 μμc/l. On May 1, after 0.3 inches of the most active rain had fallen within four days, the cistern water assayed less than 10 μμc/l.

Several samples of surface water from a small creek in the Cincinnati area were collected on May 5 and 6 following 0.32 inches of rain on May 5. The creek water activity was 69 to 163 μμc/l, and of this 41 to 51 percent was soluble. The Cincinnati tap water during this period had an activity of less than 30 μμc/l.

From these scattered tests it appears that most of the radioactivity is readily removed from the rain water by natural purification.

SUMMARY

The precipitation from March 1954 to March 1954 has been sampled and tested for suspended and soluble beta radioactivity. During and immediately following the 1953 Spring Series weapon tests (March 19 to June 29) the radioactivity in the 10.86 inches of rainfall varied

from 280 to 319,000 μμc/l as compared to a pre-bomb test background activity of 50 μμc/l. Only three rains out of ten tests in January and February 1954 showed a level of activity greater than the pre-bomb test background.

The activity of rains can generally be attributed to particular bomb bursts on the basis of a comparison of the half-lives of rain activities to the theoretical decay of fission product.

It was estimated that on June 10 and December 10, the accumulated rain activity which had fallen per square mile would have decayed to about 2.4 and 0.15 curies, respectively.

Surface and cistern waters collected following rains having an activity of 50 to 500 times background showed an activity within three times background.

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Phys. Rev. 73, 1318 (1948).

Table 1

RADIOACTIVITY FALL-OUT IN RAIN IN THE CINCINNATI AREA

Sample Series	Date of Bomb Burst	Precipitation*		Radioactivity			
		Date Ending		Total		Soluble	
			In.	μμc/ml	mc/sq.mi.	μμc/ml	Percent
0		3-2-53	0.228	0.05	0.73	0.03	60
		3-4	0.70	0.03	1.40	0.03	100
		3-5	0.198	0.08	1.00	0.02	25
		3-9	0.058	0.04	0.13	0.02	50
		3-12	0.21	0.04	0.55	0.02	50
		3-16	0.45	0.06	1.76	0.04	67
		3-18	0.67	0.06	2.70	0.04	67
1	3-17						
		3-19	0.03	1.60	3.2	0.24	15
		3-23	0.02	2.50	3.8	0.98	39
2	3-24						
		3-31	0.14	3.66	33.7	0.17	5
		3-31b	0.04	0.28	0.7	0.08	28
3	3-31						
		4-1	0.21	2.32	33.6	0.34	15
		4-1b	0.09	2.74	18.0	0.61	22
		4-6	0.20	3.82	40.3	0.91	24
		4-6b	0.10	2.00	13.2	0.56	28
		4-7	0.08	1.39	5.2	0.49	35

Table 1 (Continued)

Sample Series	Date of Bomb Burst	Precipitation*		Radioactivity			
				Total		Soluble	
		Date Ending	In.	μμc/ml	mc/sq.mi.	μμc/ml	Percent
4	4-6						
		4-10	0.20	2.30	30.2	0.28	12
		4-13	0.20	2.83	35.2	0.67	24
		4-15	0.06	2.91	11.7	0.43	15
		4-15a	0.12	2.57	20.6	0.95	37
5	4-11						
		4-16	0.18	6.89	81.5	1.09	16
		4-18	1.00	2.59	173.	0.26	10
		4-18b	0.13 S	0.27	2.3	0.03	11
6	4-18						
		4-19a	0.08	0.79	4.2	0.04	5
		4-19b	0.01	2.69	1.8	0.13	5
		4-23	0.01	2.11	1.4	0.00	0
		4-27	0.20	1.76	23.4	0.33	19
7	4-25						
		4-28	0.01	31.1	20.7	22.7	73
		4-29	0.02	319.	426.	95.7	30
		4-30	0.08	96.3	512.	11.4	12
		4-30b	0.01	21.2	14.3	-	-
		5-1	0.22	13.3	198.	-	-
		5-5	0.32	7.43	156.	0.82	11
		5-8	0.26	1.70	30.	0.55	32

Table 1 (Continued)

Sample Series	Date of Bomb Burst	Precipitation*		Radioactivity			
		Date Ending	In.	Total		Soluble	
				μμc/ml	mc/sq.mi.	μμc/ml	Percent
8	5-8						
		5-11b	0.06	2.85	11.5	0.04	1
		5-12	0.02	0.95	1.2	-	
		5-12b	0.09	4.09	24.2	1.52	37
		5-13	0.08	3.70	19.5	1.17	32
		5-13a	0.08	3.23	17.0	0.48	15
		5-13b	0.11	1.81	13.0	0.41	23
		5-14	0.04	2.14	5.6	0.65	30
		5-15	0.22	1.61	23.	0.54	34
		5-16b	0.86	1.21	68.	0.30	25
9	5-19						
		5-20	0.02	3.14	4.2	0.32	10
		5-22	1.55	85.7	8750.	2.34	27
		5-24	0.28	75.1	1380.	8.61	11
		5-25	0.09	30.3	180.	19.7	65
10	5-25						
		6-7a	1.32	2.71	235.	0.63	23
11	6-4						
		6-10	0.03	20.6	40.6	16.0	78
		6-10b	0.71	1.84	860.	0.46	25
12	-						
		6-15	0.12	1.64	12.9	1.63	100
		6-17	0.35	1.54	35.4	0.37	24
		6-29	0.42	1.80	49.6	0.12	7

Table 1 (Continued)

Sample Series	Date of Bomb Burst	Precipitation*		Radioactivity			
				Total		Soluble	
		Date Ending	In.	μμc/ml	mc/sq.mi.	μμc/ml	Percent
13	-						
		7-2	0.65	0.17	7.3	0.03	16
		7-6b	1.12	0.23	17.2	0.06	27
		7-7a	0.02	0.40	0.5	0.17	42
		7-17a	1.30	0.16	14.0	0.08	50
		7-20	0.42	0.24	6.8	0.15	60
		7-22a	0.29	0.54	10.0	0.26	50
		7-24	0.17	0.09	0.9	0.05	63
14	-						
		8-3	0.24	0.14	2.1	0.04	30
		8-4	0.51	0.05	1.8	0.03	50
		8-5	0.05	0.04	0.1	0.02	37
		8-6	0.01	0.03	0	0.01	41
		8-10	0.22	0.19	2.7	0.06	30
		8-17a	0.02	0.39	0.6	0.11	29
15	-						
		9-8	0.07	2.69**	-	2.35**	87
		10-4	0.05	0.43	1.4	0.28	66
		10-27	0.38	0.08	2.0	0.08	100
		11-23	0.90	0.07	4.1	0.06	86
		11-25	0.20	0.06	0.8	0.05	83
		11-27	0.15	0.11	1.1	0.08	73
		11-30	0.01	0.17	0.1	0.16	94

Table 1 (Continued)

Sample Series	Date of Bomb Burst	Precipitation*		Radioactivity			
				Total		Soluble	
		Date Ending	In.	μμc/ml	mc/sq.mi.	μμc/ml	Percent
16	-						
		12-4	0.05	0.11	0.36	0.03	27
		12-9	0.03	0.09	0.18	0.07	78
		12-14	0.67	0.06	2.64	0.06	100
		12-14a	0.15	0.09	0.90	0.08	89
		12-14b	0.16	0.11	1.2	0.09	82
		12-15	0.01	0.07	0.05	0.07	100
		12-16	0.01	0.07	0.05	0.04	57
		12-21	0.08	0.12	0.63	0.08	67
		12-23	0.04	0.08	0.21	0.06	75
		12-28	0.16	0.10	1.05	0.08	80
17	-						
		1-2-54	0.01	0.05	0.03	0.05	100
		1-9b	0.39	0.06	1.54	0.06	100
		1-15b	0.35	0.03	0.69	0.01	33
		1-18b	0.36	0.00	0.00	0.00	0
		1-19b	0.03	0.13	0.26	0.09	69
		1-20	0.36	0.01	0.24	0.01	100
		1-26b	0.01	0.05	0.03	0.04	80
18	-						
		2-4	0.01	0.02	0.01	0.02	100
		2-24	0.02	0.08	0.10	0.03	38
		2-26	0.018	0.17	0.11	0.06	35

Notes:

* Unless otherwise indicated, the collections were made between 0800 and 0900. Dates followed by the letters a and b indicate collections were made at 1130-1230, and at 1600-1630, respectively. A few rain samples in October and several in January and in February were not sampled.

** Laboratory contamination suspected.

S Melted snow.

Table 2

HALF-LIFE OF RAIN SAMPLES OF MAXIMUM ACTIVITY BETWEEN BOMB BURSTS

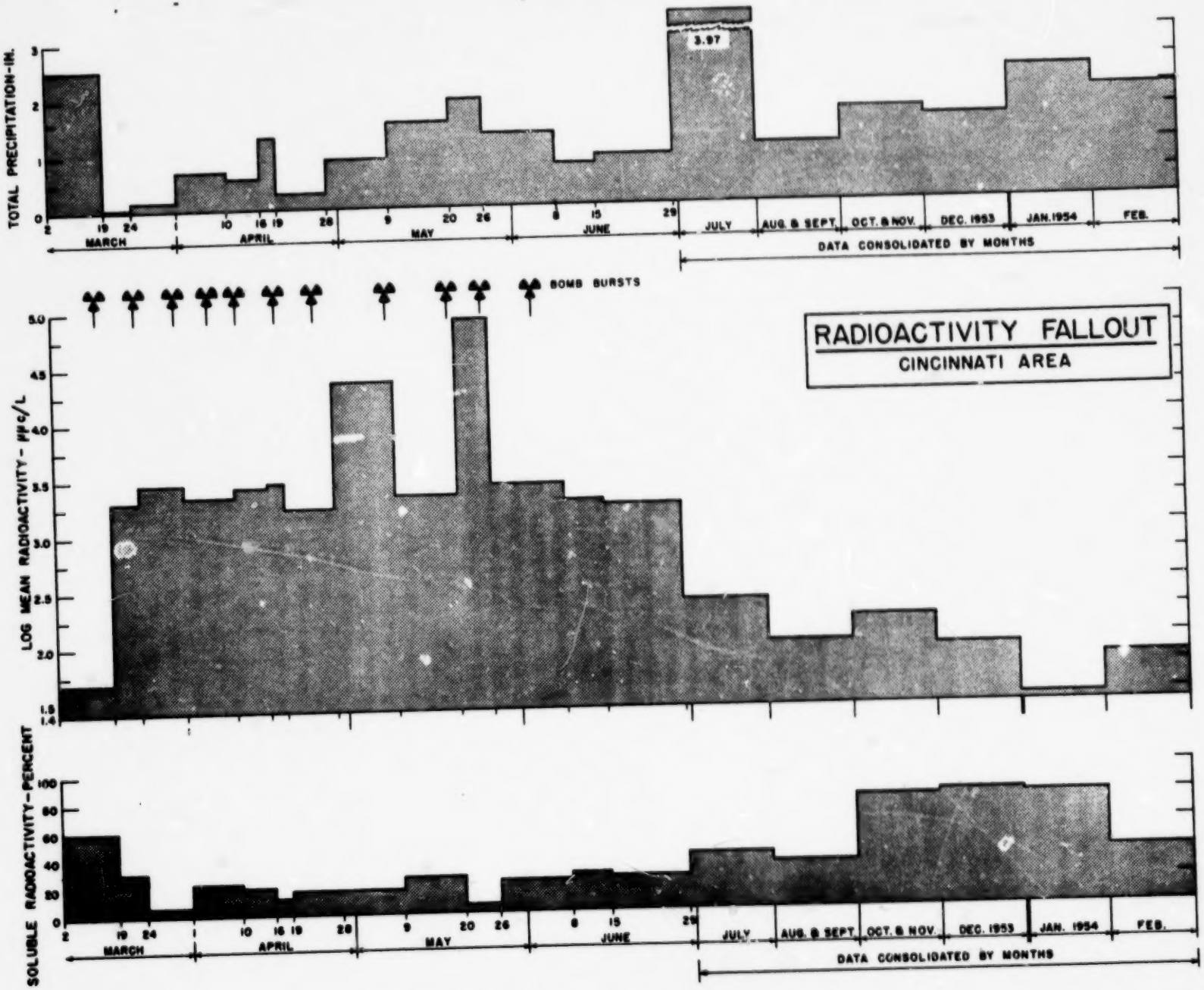
Sample Series	Sample Date	Initial Decay Date	Half-Life of Rain Days	Half-Life (in days) for Fission Product formed on										
				3-17	3-24	3-31	4-6	4-11	4-18	4-25	5-8	5-19	5-25	6-4
				Bomb Burst Number										
				1	2	3	4	5	6	7	8	9	10	11
1	3-19	3-20	3.2	[2.3]										
2	3-31	4-1	4.5	11.3	[6.2]									
3	4-6	4-6	5.0		9.7	[4.7]								
4	4-13	4-13	14.		[15.]	9.7	5.5							
5	4-16	4-17	3.7				8.3	[4.7]						
6	4-19b	4-20	9.0				[10.5	7.0]	1.5					
7	4-29	4-29	3.4						8.3	[3.3]				
8	5-11b	5-11	8.5							[12.0	2.3]			
9	5-22	5-22	2.0								10.5	[2.3]		
10	6-7	6-8	17.								25.3	[15.0]	10.5	3.3
11	6-10	6-10	10.										[12.0	4.7]

Table 3

SUMMARY

Sample Series	Date of Bomb Burst	Average Age	Total Rain	Radioactivity			Residual Activity	
				Weighted Average	Total	Soluble	On June 10 1953	On Dec. 10 1953
		Days	In.	μμc/ml	mc/sq.mi.	Percent	mc/sq.mi.	mc/sq.mi.
0	-	-	2.49	0.05	8.3	60	-	-
1	3-17	4	0.05	2.13	7.0	31	0.18	0.04
2	3-24	7	0.18	2.90	34.4	5	1.9	0.45
3	3-31	4	0.68	2.47	110.	23	3.5	0.74
4	4-6	6.5	0.58	2.56	98.	20	6.1	1.22
5	4-11	6	1.31	2.98	257.	12	16.2	3.01
6	4-18	5	0.30	1.56	30.8	17	1.8	0.30
7	4-25	6	0.92	22.4	1360.	17	118.	16.9
8	5-8	5.5	1.56	1.78	183.	26	21.	2.22
9	5-19	3.5	1.94	80.6	10,300	5	1135	76.6
10	5-25	13	1.32	2.71	235	23	183	8.84
11	6-4	6	0.74	1.85	900	27	900	14.75
12	-	18	0.89	1.67	98	25	-	6.88
13	-	37	3.97	0.22	56.7	40	-	12.70
14	-	67	1.05	0.105	7.3	34	-	-
15	-	150	1.69	0.085	9.3	79	-	-
16	-	195	1.36	0.08	7.27	82	-	-
17	-	224	1.51	0.03	2.79	80	-	-
18	-	256	0.04	0.08	0.22	40	-	-
			Total		13,515.*		2,386.	145.

* Excluding sample series 0 and 12 to 18, inclusive.


TOTAL PRECIPITATION-IN.
3.97
MARCH
APRIL
MAY
JUNE
JULY
AUG. & SEPT.
OCT. & NOV.
DEC. 1953
JAN. 1954
FEB.
DATA CONSOLIDATED BY MONTHS
BOMB BURSTS
RADIOACTIVITY FALLOUT
CINCINNATI AREA
LOG MEAN RADIOACTIVITY - μμc/L
SOLUBLE RADIOACTIVITY - PERCENT